



High-performance organic solar cells utilizing graphene oxide in the active and hole transport layers

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ABSTRACT

We have successfully synthesized and employed graphene oxide (GO) to boost photons harvesting and charge transport process in thin film organic solar cells (TFOSCs). The graphene oxide was inlaid in both the P3HT:PCBM-based photoactive medium of the device, as well as, a dopant in PEDOT:PSS hole transport buffer layer (HTL). The parameters of the solar cells produced with the inclusion of GO in the HTL and the active layer results in high short-circuit current densities (J_{sc}), which translated into high power conversion efficiencies (PCEs). GO in the HTL facilitates charge transport, selective electron blocking and hole injection at the interface for enhanced device performance. On the other hand, the use of GO in the active layer remarkably improves the optical absorption leading to high charge carriers photogeneration requisite to efficient OSCs. Similarly, effective exciton dissociation is energetically favoured in the GO modified active layer devices which corroborated with improved conductivity of the medium that assisted charge carriers transport processes. Enhanced photocurrent has been recorded, as high as 18 mA cm^{-2} , from the TFOSCs by the inlay of GO in the active layer. Consequently, increased PCE of up to 40% and 120% is achieved by the inclusion of GO in the HTL and photoactive layers, respectively.

1. Introduction

Organic photovoltaics (OPVs) are currently under extensive research as a viable alternative to the silicon-based solar cells in solar energy harvesting. These have the advantage of low-cost solution-processability, light weight, flexibility and ease in fabrication. A typical organic solar cell (OSC) consists of the electrodes, interfacial layers and active layer which are fabricated in the bilayer or bulk heterojunction (BHJ) device architecture. The optical and electronic properties of the active layer determine the generation, separation and transport of the photoinduced charge carriers in OSCs. On the other hand, the interfacial layers serve to ensure effective extraction and transportation of the photoinduced charge carriers to the electrodes. Hence, the inherent properties of the active and interfacial layer materials are key in designing high-performance OSCs. In BHJ-OSCs active layer, the acceptor material is randomly dispersed within the donor phase creating BHJs between the acceptor nanostructure and the donor polymer matrix. The BHJs not only provide sites for exciton dissociation but also provide bicontinuous pathways for charge carrier transport (Moon et al., 2008; Wan et al., 2011). However, the BHJ-solar cells suffer from misaligned

donor and acceptor energy levels, small exciton diffusion length, low optical absorption and low charge carrier mobility (Vandewal et al., 2009). Of these factors, low carrier mobility is the prevalent cause of poor performance in BHJ-OSCs as it results in charge carrier recombination and/or trapping before getting to the electrodes (O'Malley et al., 2012). The interfacial layers serve to inhibit charge carrier recombination by the selective and efficient transportation of holes and electrons to the respective electrodes (Murase and Yang, 2012). Hence, a suitable device architecture, and an appropriate donor and acceptor materials would be key to efficient photoinduced charge carrier generation, separation and transportation. Likewise, an efficient interfacial layer would be determinant in the extraction and transportation of the charge carriers to the electrodes to yield a high-efficiency OSC.

Currently, the active layer of some of the top-performance OSCs are based on polymer-fullerene blends, in which the commonly used donor and acceptor materials are poly-3-hexylthiophene (P3HT) and (6–6) phenyl-C61-butyric acid methyl ester (PCBM), respectively. The polymer, P3HT can self-assemble into a well-ordered semi-crystalline structure which enables π - π stacking of its conjugated backbone thus, leading to an improved charge carrier mobility (Yamamoto et al.,

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1998). Also, P3HT exhibits a high optical absorption coefficient of $ca. 10^5 \text{ cm}^{-1}$ (Miller et al., 2008). Besides, P3HT is relatively stable (Jørgensen et al., 2012) and is viable for large-area roll-to-roll processing (Espinosa et al., 2014) making it suitable in commercial OPVs production. On the other hand, the fullerene acceptor PCBM, is associated with poor optical absorption, difficulty in band gap engineering and morphological instability (Kim et al., 2015b). An ideal energy level alignment at the electrodes-active layer interfaces is essential for maximized open-circuit voltage (V_{oc}) hence, good device performance in BHJ-OSCs. To this end, the semiconducting polymer poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) is usually used as a hole transport layer (HTL) in OSCs. PEDOT:PSS has a high work function value $ca. 5.2 \text{ eV}$ matching the highest occupied molecular orbital (HOMO) level of most donor polymers. Thus, it ensures good electrical contact at the anode-active layer interface. In the typical BHJ-OSCs, PEDOT:PSS aligns the energy levels of P3HT and the indium tin oxide (ITO) anode and also smoothens the rough surface of ITO (van Reenen et al., 2014). Nevertheless, the high acidity and hygroscopic nature of PEDOT:PSS impairs its long-term stability. Moreover, PEDOT:PSS suffers from a not only inhomogeneous but also a relatively low electrical conductivity because of the insulating PSS chains (Hu et al., 2016; Kim et al., 2015a).

Because of the aforementioned shortcomings in the BHJ-OSCs, carbon based materials such as carbon nanotubes and graphene which are characterized by high carrier mobility have been employed in the active and interfacial layers to provide percolation pathways for charge transport. Graphene, a 2-D nanomaterial, consists of a monolayer of sp^2 -hybridized carbon atoms in a honeycomb lattice structure. The outstanding electrical and optical properties of graphene coupled with the solution-based processability and availability in its functionalized forms render it suitable for application in OPV. Furthermore, graphene has the advantage of thermal stability and high specific surface area. These properties make graphene viable for application as transparent conductive electrode (Konios et al., 2015), cathode, interfacial layer and active layer materials in OSCs (Dai, 2012; Iwan and Chuchmała, 2012; Manzano-Ramírez et al., 2015; Singh and Nalwa, 2015). Graphene oxide (GO), a derivative of graphene, comes in handy in modifying the active layer and HTL of BHJ-OSCs as it enables functionalization of graphene's carbon backbone, and simple low-cost device fabrication elementary in OPV technologies. GO is an oxygen functionalized graphene sheet consisting of oxygen functional groups such as the epoxy and hydroxylic groups on the basal planes while the carboxylic group are mainly at the edges (Dreyer et al., 2010). The carboxylic group carbon atoms are bonded to carbon or oxygen atoms in a sp^2 -hybridized system (Eda et al., 2010). On the other hand, the epoxy and hydroxyl

groups carbon atoms which are sp^3 -hybridized are bonded to oxygen atoms (Yang et al., 2009). Because of these oxygen functional groups, GO is characterized by a disrupted sp^2 conjugation of the graphitic lattice thus, effecting insulating behaviour. Charge transport in GO is via hopping within the localized sp^2 sites (Eda et al., 2009). Thus, in a BHJ-OSCs modified by GO, charge carriers' injection, and transportation can easily occur through the localized sp^2 sites.

In this study, we report the use of GO in the modification of the HTL and active layer of BHJ-OSCs. GO was dispersed in PEDOT:PSS and P3HT:PCBM and used as the HTL and photoactive layer of BHJ-OSCs, respectively. The OPV devices were characterized for photovoltaic performance and the results are discussed in details. Further, a control device with PEDOT:PSS as HTL and P3HT:PCBM as active layer was characterized for comparison purposes. This study shows that the usage of GO in the active layer can significantly improve the optical absorbance and subsequently the J_{sc} , which directly correlates to the ultimate PCE of the devices. GO has the advantage of solubility in organic solvents thus, affords ease in solution-processability of OSCs. Owing to the 2-D planar structure, it is envisaged that GO would form wide donor-acceptor interfaces in the active layer for photoinduced charge generation, and would create percolating pathways for the electrons transport. Besides, it has optical properties and work function comparable to PEDOT:PSS thus, would be a suitable HTL material (Li et al., 2010). Studies on the utilization of GO as a HTL of OSC (Li et al., 2010; Stratakis et al., 2013; Stratakis et al., 2014; Konios et al., 2016) and in the modification of the PEDOT:PSS HTL of a perovskite solar cell (Li et al., 2016) have been reported. Similarly, GO has been used as the electron acceptor material in the active layer of OSCs (Bkakrai et al., 2015).

2. Experimental section

2.1. Materials

Graphite powder ($< 150 \mu\text{m}$, 99.99%) was bought from Sigma-Aldrich South Africa. Sodium nitrate (99%) and potassium permanganate (99%) were purchased from Ace, South Africa while 98% sulphuric acid was purchased from Promark Chemicals, South Africa. Hydrogen peroxide (30%) was bought from Merck, South Africa. PEDOT:PSS (M124) and PC₆₀BM (95%) were acquired from Ossila ltd while P3HT (MW $> 45,000$ GPC) was purchased from Lumtec. ITO substrates were bought from Ossila ltd. These were used as obtained.

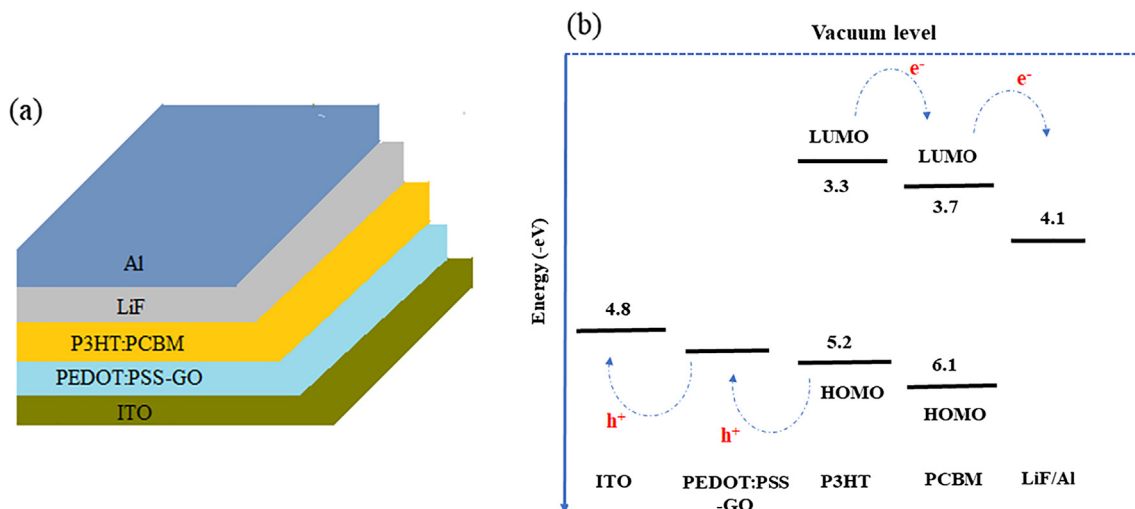


Fig. 1. (a) Schematic diagram of the GO modified HTL device and (b) energy level diagram of the device's constituent materials.

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